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14. ABSTRACT This project provided training for graduate students in a wide variety of areas related to III-nitride research: epitaxial growth using MOCVD and MBE, optical and electrical characterization, post-growth processing and device fabrication. This training program was unique in that it exposed students to nearly all aspects of critical III-nitride technology through this comprehensive training.				
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AFOSR – BMDO – AASERT Final Report

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(April 15, 1998 to October 14, 2001)

“BMDO-AASERT Group III Nitride Semiconductor nanostructure research
MOCVD Growth and Novel Characterizations of high Temperature, high carrier
density and microcrack lasing effects”

December 10, 2001

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Summary

This award provided training for several graduate students working in the laboratory of Dr. Jin-Joo Song in the Center for Laser and Photonics Research at Oklahoma State University, over the period 15 April 1998 - 14 October 2001. This award supported four graduate students; three of these completed their Ph.D. degrees and presented successful theses. One other left the laboratory and transferred to the laboratories of other OSU faculty in the College of Engineering.

The scientific training made possible through this award is evidenced by the progress described in detail in the publications outlined in this report. These publications were co-authored by one or more of the supported students during the period of the award. Difficulties with the construction and the bringing ``on-line'' of the MOCD facilities at the OSU Clean Room required that most of the scientific experiments and analyses were performed on materials and devices grown and constructed by collaborators from other institutions (identified in the publications).

The delays with the Clean Room and MOCVD facilities have been previously described in a letter from the PI (Dr. Song) to Dr. Johnstone (18 April 2000). With reference to that letter, a second MOCVD unit has now been installed; stimulated emission and optical and electrical characterization of advanced nitride structures were performed; and femtosecond laser-induced carrier density experiments and theoretical analysis to interpret the carrier relaxation phenomena were carried out.

The attached data describes in detail the scientific work and the progress of the supported students.

Students Supported

Student Name/ Grant Position	Degree (Ph.D., M.S., B.S.)	Date Supported by Grant	Graduation Date (if applicable)	Currently
Gordon H. Gainer/ Graduate Research Assistant	Ph.D., Physics	07-01-98 to 07-01-01	July 2001	Working in the laboratory of Dr. Jerzy Krasinski, Dept of Electrical & Computer Engineering, Oklahoma State University
Jack Biu Lam/ Graduate Research Assistant	Continuing Student	07-01-98 to 10-14-01	N/A	Transferred to Dr. Krasinski's laboratory
Brian D. Little/ Graduate Research Assistant	Ph.D., Physics	07-01-99 to 12-31-00	December 2000	Chicago, IL
Theodore Schmidt/ Graduate Research Assistant	Ph.D., Physics	07-01-98 to 12-31-98	December 1998	Unknown

Name: Gordon H. Gainer

Date of Degree: August, 2001

Institution: Oklahoma State University

Location: Stillwater, Oklahoma

Title of Study: EMISSION MECHANISMS OF InGaN-BASED III-NITRIDE HETEROSTRUCTURES

Pages in Study: 216

Candidate for the Degree of Doctor of Philosophy

Major Field: Physics

Scope and Method of Study: The emission mechanisms of InGaN-based III-nitride heterostructures were determined to aid in the design of InGaN-based light emitting diodes and laser diodes. $In_{0.18}Ga_{0.82}N$ epilayers and $In_{0.18}Ga_{0.82}N/GaN$ MQWs were studied by photoluminescence, photoluminescence excitation, time resolved photoluminescence, absorption, and stimulated emission. These measurements were performed as a function of excitation power density, excitation photon energy, excitation length, and temperature.

Findings and Conclusions: The photoluminescence peak energy had an "S-shaped" temperature dependence (redshift, blueshift, and then redshift with increasing temperature). Initially, with increasing temperature above 10 K, the charge carrier decay time increases, indicating the dominance of radiative recombination. This gives the carriers more opportunity to relax to lower energy band tail states before radiatively recombining, and this causes the initial redshift. With further temperature increases, the decay time decreases due to nonradiative processes, and this causes the blueshift. Then, with further temperature increases, the regular bandgap shrinkage causes a redshift, which is not as great as it would be without the band filling. The localized band tail states are caused by defects, such as In alloy inhomogeneity and layer thickness variations. These defects enhance the spontaneous and stimulated emission by localizing carriers and preventing them from migrating to nonradiative recombination centers. A mobility edge was found to be well above the spontaneous and stimulated peak positions, but much lower than the absorption edge, indicating that both the spontaneous and stimulated emissions originate from deep carrier localization. Carrier localization was also shown to be the spontaneous and stimulated emission mechanism by much more evidence, such as a strong correlation between the inverse stimulated emission threshold density and photoluminescence excitation spectra. Epitaxial lateral overgrowth was described as a method to greatly reduce the density of harmful defects, while keeping the relatively beneficial defects of bandgap inhomogeneity.

ADVISER'S APPROVAL: _____

Name: Brian Dean Little

Date of Degree: December, 2000

Institution: Oklahoma State University

Location: Stillwater, Oklahoma

Title of Study: **OPTICAL PROPERTIES OF WIDE BANDGAP III-NITRIDE- AND ZINC OXIDE-BASED EPILAYERS, ALLOYS, AND HETEROSTRUCTURES**

Pages in Study: 179

Candidate for the Degree of Doctor of Philosophy

Major Field: Physics

Scope and Method of Study: The optical properties of the III-Nitrides and ZnO were studied using a variety of spectroscopic experimental techniques. The semiconductor samples studied included epilayers, bulk crystals, alloys, and heterostructures. A large variety of picosecond and nanosecond laser systems as well as continuous wave light sources were used to optically excite the samples in this study. The sample emission was detected with photomultiplier tubes, CCD cameras, and optical multi-channel analyzers in conjunction with spectrometers.

Findings and Conclusions: The binding energy for intrinsic excitons in GaN was determined. The effect of strain on these excitons was studied. The optical properties of InGaN alloys were investigated. The characteristics of AlGaN and InGaN alloys as a function of pressure were studied. The dynamical behavior of photoexcited carriers in AlGaN/GaN double heterostructures was explained. The stimulated emission from AlGaN/GaN separate confinement heterostructures was studied as a function of the excitation wavelength. A comparison of the emission from highly-excited (In,Al)GaN thin films and heterostructures was performed. The results obtained suggest that the group-III Nitrides and ZnO are excellent candidates for the development of ultraviolet optoelectronic devices. Several important optical parameters are provided, which are critical for understanding and designing efficient devices.

ADVISER'S APPROVAL: _____

Name: Theodore J. Schmidt

Date of Degree: December, 1998

Institution: Oklahoma State University

Location: Stillwater, Oklahoma

Title of Study: OPTICAL SPECTROSCOPY OF HIGHLY EXCITED GROUP III
NITRIDES

Pages in Study: 235

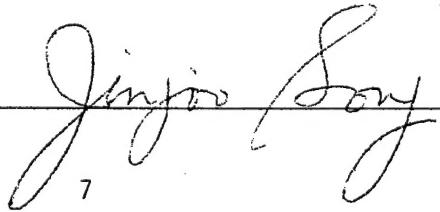
Candidate for the Degree of Doctor of Philosophy

Major Field: Physics

Scope and Method of Study: The optical properties of highly excited group III nitride semiconductors was studied. The research was undertaken to gain a better understanding of the optical phenomena associated with stimulated emission and lasing in this wide band gap semiconductor system. The optical properties of AlGaN, GaN, InGaN, and their related heterostructures were monitored as the number of photo-generated free carriers was increased beyond that required to achieve population inversion. Experimental techniques used include: Optically pumped stimulated emission studies, variable-stripe gain spectroscopy, energy selective optical excitation spectroscopy, and nondegenerate optical pump-probe spectroscopy. Particular emphasis is placed on the optical properties of highly excited InGaN/GaN multiple quantum well structures.

Findings and Conclusions: Stimulated emission and lasing was observed from all materials studied. Stimulated emission was observed at temperatures exceeding 700 K, indicating this material system is particularly well suited for high temperature opto-electronic applications. The mechanisms leading to optical gain in each material was ascertained as a function of temperature and optical excitation intensity. The stimulated emission characteristics of AlGaN were found to be similar to that of GaN, while InGaN exhibited markedly different behavior. The results of nondegenerate optical pump-probe experiments indicate the group III nitrides are well suited for optical switching applications.

ADVISOR'S APPROVAL:


Jin-Joo Song

Publications List

1. "Femtosecond pump-probe spectroscopy and time-resolved photoluminescence of an $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$ double heterostructure," C.K. Choi, B.D. Little, Y.H. Kwon, J.B. Lam and J.J. Song, *Physical Review B* **63**, 195302 (2001).
2. "Well-thickness dependence of emission from GaN/AlGaN separate confinement heterostructures," G.H. Gainer, Y.H. Kwon, J.B. Lam, S. Bidnyk, A. Kalashyan, J.J. Song, S.C. Choi and G.M. Yang, *Applied Physics Letters* **78**, 3890 (2001).
3. "Optical properties and lasing in $(\text{In},\text{Al})\text{GaN}$ structures," S. Bidnyk, G.H. Gainer, S.K. Shee, J.B. Lam, B.D. Little, T. Sugahara, J. Krasinski, Y.H. Kwon, G.H. Park, S.J. Hwang, J.J. Song, G.E. Bulman, H.S. Kong, *Phys. Stat. Sol. (a)* **183**, 105 (2001).
4. "Linear and nonlinear optical properties of $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$ heterostructures," Y.H. Cho, T.J. Schmidt, S. Bidnyk, G.H. Gainer, J.J. Song, S. Keller, U.K. Mishra and S.P. DenBaars, *Physical Review B* **61**, 7571 (2000).
5. "Dynamics of anomalous optical transitions in $\text{Al}_x\text{Ga}_{1-x}\text{N}$ alloys," Y.H. Cho, G.H. Gainer, J.B. Lam, J.J. Song, W. Yang, W. Jhe, *Physical Review B* **61**, 7203 (2000).
6. "Time-resolved study of yellow and blue luminescence in Si-and Mg-doped GaN ," Y.H. Kwon, S.K. Shee, G.H. Gainer, G.H. Park, S.J. Hwang and J.J. Song, *Applied Physics Letters* **76**, 840 (2000).
7. "Study of gain mechanisms in AlGaN in the temperature range of 30-300 K," J.B. Lam, S. Bidnyk, G.H. Gainer, B.D. Little and J.J. Song, *Applied Physics Letters* **77**, 4101 (2000).
8. "MOCVD growth, stimulated emission and time-resolved PL studies of $\text{InGaN}/(\text{In})\text{GaN}$ MQWS: Well and barrier thickness dependence," S.K. Shee, Y. Kwon, J. Little, J.B. Lam, G.H. Gainer, G.H. Park, S.J. Hwang, J.J. Song, *Journal of Crystal Growth* **221**, 373 (2000).
9. "Comparative study of near-threshold gain mechanisms in GaN epilayers and GaN/AlGaN separate confinement structures," S. Bidnyk, J.B. Lam, B.D. Little, G.H. Gainer, Y.H. Kwon, J.J. Song, *The International Society for Optical Engineering (SPIE) Conf. Proc.* **3947**, 126 (2000).
10. "Microcavity-based semiconductor lasers for near- and deep-UV applications," S. Bidnyk, J.B. Lam, B.D. Little, Y.H. Kwon, J.J. Song, G.E. Bulman, H.S. Kong, *Conference on Lasers and Electro-Optics (CLEO) 2000 Technical Digest*, CMG5 (2000).
11. "GaN/AlGaN SCH UV semiconductor lasers: Effect of GaN well thickness on lasing efficiency," G.H. Gainer, Y.H. Kwon, J.B. Lam, A. Kalashyan, J.J. Song, S.C. Choi, G.M. Yang, *Conference on Lasers and Electro-Optics (CLEO) 2000 Technical Digest*, CMG4 (2000).
12. "A study of the structural and optical properties of $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$ quantum wells with different In compositions," Y.H. Kwon, G.H. Gainer, S. Bidnyk, Y.H. Cho, J.J. Song, M. Hansen, S.P. DenBaars, *Mat. Res. Soc. Symp. Proc.* **595**, MRS Internet J. Nitride Semicond. Res. **5S1**, W12.7 (2000).

13. "Dynamics of anomalous temperature-induced emission shift in MOCVD-grown (Al, In)GaN thin films," Y.H. Cho, G.H. Gainer, J.B. Lam, J.J. Song, W. Yang, W. Jhe, Mat. Res. Soc. Symp. Proc. **595**, MRS Internet J. Nitride Semicond. Res. **5S1**, W11.57 (2000).
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17. "Optical properties of In_xGa_{1-x} alloys grown by metalorganic chemical vapor deposition," W. Shan, W. Walukiewicz, E.E. Haller, B.D. Little, J.J. Song, M. McCluskey, N. Johnson, Z. Feng, M. Schurman, R. Stall, Journal of Applied Physics **84**, 4452 (1999).
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20. "Carrier dynamics of abnormal temperature-dependent emission shift in MOCVD-grown InGaN epilayers and InGaN/GaN quantum wells," Y.H. Cho, B.D. Little, G.H. Gainer, J.J. Song, S. Keller, U.K. Mishra, S.P. DenBaars, MRS Internet J. Nitride Semicond. Res. **4S1**, G2.4 (1999).
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25. "Critical issues of localization in the development of InGaN/GaN laser diodes," S. Bidnyk, Y.H. Cho, T.J. Schmidt, G.H. Gainer, J.J. Song, S. Keller, U.K. Mishra, S.P. DenBaars,

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32. "Ultrafast carrier dynamics in GaN epilayers studied by femtosecond pump-probe spectroscopy," A.J. Fischer, B.D. Little, T.J. Schmidt, C.K. Choi, J.J. Song, The International Society for Optical Engineering (SPIE) Conf. Proc. 3624, 179 (1999).
33. "Time-resolved photoluminescence studies of GaN, InGaN, and AlGaN grown by metalorganic chemical vapor deposition," Y.H. Cho, G.H. Gainer, J.J. Song, S. Keller, U.K. Mishra, S.P. DenBaars, W. Yang, S.A. McPherson, The International Society for Optical Engineering (SPIE) Conf. proc. 3624, 283 (1999).
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35. "Optical characteristics of group III nitride quantum structures," Y.H. Cho, W. Jhe, T.J. Schmidt, S. Bidnyk, G.H. Gainer, J.J. Song, Proceedings of the 3rd Korea-China Joint Workshop on Advance Materials (invited), 351 (1999).
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37. "Comparison of spontaneous and stimulated emission from UV-blue photonics materials," B.D. Little, Y.H. Cho, T.J. Schmidt, G.H. Gainer, J.B. Lam, J.J. Song, W. Yang, S. Keller, U.K. Mishra, S.P. DenBaars, W. Jhe, Conference on Laser and Electro-Optics, CLEO/Pacific Rim '99, Technical Digest **2**, 290 (1999).
38. "Nonlinear optical spectroscopy of highly excited InGaN/GaN multiple quantum well blue laser structures," T.J. Schmidt, S. Bidnyk, Y.H. Cho, G.H. Gainer, J.J. Song, 1999 CLEO Technical Digest Series, **Ctu4**, 202 (1999).
39. "A novel technique to evaluation of optical confinement in semiconductor laser structures through spatially and spectrally resolved emission spectra," S.Bidnyk, T.J. Schmidt, B.D. Little, J. Krasinski, J.J. Song, 1999 CLEO Technical Digest Series, **CtuK52**, 145 (1999).
40. "Carrier recombination dynamics of $Al_xGa_{1-x}N$ epilayers grown by MOCVD," Y.H. Cho, G. H. Gainer, J.B. Lam, J.J. Song, W. Yang, S.A. McPherson, Mat. Res. Soc. Symp. Proc., **572**, 457 (1999).
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44. "Intrinsic exciton transitions in GaN," W. Shan, A.J. Fischer, S.J. Hwang, B.D. Little, R. Hauenstein, X.C. Xie, J.J. Song, D. Kim, B. Goldenberg, R. Horning, S. Krishnankutty, W. Perry, M. Bremer, R. Davis, Journal of Applied Physics **83**, 455 (1998)
45. "Laser action in GaN pyramids grown on (111) silicon by selective lateral overgrowth," S. Bidnyk, B.D. Little, Y. Cho, J. Krasinski, J.J. Song, W. Yang, S. McPherson, Applied Physics Letters **73**, 2242 (1998).
46. "S-shaped temperature-dependent emission shift and carrier dynamics in InGaN/GaN multiple quantum wells," Y. Cho, G.H. Gainer, A. Fischer, J.J. Song, S. Keller, U. Mishra, S. DenBaars, Applied Physics Letters **73**, 1370 (1998).
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48. "High-temperature stimulated emission in optically pumped InGaN/GaN multiquantum wells," S. Bidnyk, T.J. Schmidt, Y.H. Cho, G.H. Gainer, J.J. Song, S. Keller, U.K. Mishra, S.P. DenBaars, Applied Physics Letters **72**, 1623 (1998).

49. "Pump-probe spectroscopy of band tail states in metalorganic chemical vapor deposition grown InGaN," T.J. Schmidt, Y.H. Cho, G.H. Gainer, J.J. Song, S. Keller U.K. Mishra, S.P. DenBaars, *Applied Physics Letters* **73**, 1892 (1998).
50. "High-temperature stimulated emission studies of MOCVD-grown GaN films," S. Bidnyk, B.D. Little, T.J. Schmidt, J. Krasinski and J.J. Song, *SPIE 3419*, 35 (1998).
51. "Near-band-edge photoluminescence emission in $\text{Al}_x\text{Ga}_{1-x}\text{N}$ under high pressure," W. Shan, J.W. Ager III, W. Waluiewicz, E.E. Haller, B.D. Little, J.J. Song, M. Schurman, Z.C. Feng, R.A. Stall, B. Goldenberg, *Appl. Phys. Lett.* **72**, 2274 (1998).
52. "High-temperature stimulated emission in optically pumped InGaN/GaN multi-quantum wells," S. Bidnyk, T.J. Schmidt, Y.H. Cho, G.H. Gainer, J.J. Song, S. Keller, U.K. Mishra, S.P. DenBaars, *Appl. Phys. Lett.* **72**, 1623 (1998).

Conference Presentations

1. "Optical properties and lasing in (In,Al)GaN-based structures," J.J. Song, S. Bidnyk, J.B. Lam, G.H. Gainer, and Y.H. Kwon, ISPA'2000 (invited).
2. "Study of gain mechanisms in $Al_xGa_{1-x}N$ in the temperature range of 30 to 300 K," J.B. Lam, S. Bidnyk, G.H. Gainer, B.D. Little, J.J. Song, and W. Yang, CLEO'2000, CMG1, San Francisco, CA (May 7-12, 2000).
3. "Microcavity-based semiconductor lasers for near- and deep-UV applications," S. Bidnyk, J.B. Lam, B.D. Little, J.J. Song, G.E. Bulman, and H.S. Kong, CLEO'2000, CMG5, 78, San Francisco, CA (May 7-12, 2000).
4. "GaN/AlGaN SCH UV semiconductor lasers: Effect of GaN well thickness on lasing efficiency," G.H. Gainer, Y.H. Kwon, J.B. Lam, A. Kalashyan, J.J. Song, S.C. Choi and G.M. Yang, CLEO'2000, CMG4, San Francisco, CA (May 7-12, 2000).
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ASSERT Report

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“Group III-Nitride Semiconductor Nanostructure Research: MOCVD Growth and
Novel Characterizations of High Temperature, High Carrier Density and
Microcrack Lasing Effects”

September 1997-June 2001

Through this AASERT program, graduate students were trained in the area of material growth, optical and electrical investigations of wide gap III-nitride heterostructures. Students received training on experimental apparatus as shown in the following pages, including stimulated emission (SE), photoreflectance (PR), photoluminescence excitation (PLE), and pump-probe experiments. The goal of the program was to study optical, electrical properties, epitaxial growth and photonic device applications. Blue and UV semiconductor lasers and optical modulators were of particular interest to us. During the course of the program, students actively collaborated with theoretical, MBE and MOCVD research groups from University of Illinois (Prof. Y. C. Chang) University of California at Santa Barbara (Prof. S. DenBaar), University of California at San Diego(Prof. C. Tu), Honeywell, University of North Carolina (Prof. R. F. Davis), EMCORE and CREE.

The students in our group hold many world records in important III-nitride research. These include:

- Highest temperature (700 K) lasing demonstration not only in GaN, but in any semiconductor.
- First demonstration of lasing in GaN.
- First demonstration of < 330 nm, AlGaN UV stimulated emission.
- First clear elucidation of lasing mechanisms in separate confinement heterostructures (SCHs).
- First demonstration of well defined laser fringe modes associated with internal microcracks.
- First comparison between HVPE and MOCVD GaN stimulated emission thresholds.
- MOCVD growth and first systematic studies of barrier-width dependent stimulated emission in InGaN/GaN multi-quantum-wells (MQWs).
- First femtosecond high carrier density pump-probe studies of GaN with an excitation wavelength significantly higher than the band edge.

Each subject above led to publications in reputable journals, and four invited book chapters were generated from the results. Additionally, students made numerous presentations at national and international conferences; the abstracts of refereed publication and conference presentations resulting from the AASERT award follow this report.

We firmly believe that our comprehensive and systematic growth and characterization work in III-nitrides will help understand the underlying mechanisms in lasing and hence help develop low threshold lasing structures which are more frequency agile. We have really picked up research momentum lately with progress in MOCVD growth, and consequently, there are many on-going projects which became active and effective due to this AASERT program.

Our original goals of this ASSERT program has not been changed and we are still working on the very issues of GaN related materials and related devices such as:

- MOCVD growth of high quality nitride alloy quantum structures for efficient UV-visible light emitters, including laser diodes; MOCVD growth of n-type and p-type nitride samples for various device fabrication and spectroscopic studies, including DLTS.

- Stimulated emission and optical and electrical characterization studies of the advanced nitride structures grown on campus and also at collaborating laboratories.
- Development of prototype devices using our cleanroom processing facilities, including LEDs and detectors.

We installed much equipment for device fabrication, including sub-micron lithography, plasma enhanced chemical vapor deposition, and inductively coupled plasma - reactive ion etching. We also installed much state-of-the-art equipment for optical, structural, and electrical characterization. We finished testing and adjusting this equipment to make it fully operational. We have had initial success for InGaN/GaN MQW blue LEDs. We have grown GaN, InGaN, AlGaN and their heterostructures with the goal of optimizing laser diode structures. We grew series of InGaN/GaN MQWs to find the best well and barrier thicknesses, indium concentrations, growth interruption times, etc.

In addition to studying our samples we grew by MOCVD, we investigated samples grown by hydride vapor phase epitaxy (HVPE) by other research groups. Technologies and Devices, Inc. provided us with $\text{Al}_x\text{Ga}_{1-x}\text{N}$ structures grown by HVPE on sapphire and SiC substrates. Lincoln Laboratory sent us GaN grown by HVPE. We also worked with Eagle-Picher on the surface treatment of HVPE GaN.

Additional technical details can be found in the following documents.

- A. Publication List.
- B. Dr. Brian Little's Ph.D. Thesis
- C. Dr. Gordon Gainer's Ph.D Thesis

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